PHYSICAL REVIEW B VOLUME 29, NUMBER 3 1 FEBRUARY 1984

Brief Reports

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5.4-GHz magnetic resonance of $RbMnF_3$ near T_N

D. W. Olson and R. A. Kroeger*
Department of Physics, University of Northern Iowa, Cedar Falls, Iowa 50614
(Received 14 October 1983)

The temperature dependence of the 5.4-GHz electron magnetic resonance linewidth of $RbMnF_3$ for temperatures just above and just below T_N is reported for two magnetic field directions. The $T > T_N$ linewidth variation is shown to be similar to the 9.2-GHz variation previously reported by Gupta and Seehra. It is concluded that the 5.4- and 9.2-GHz data both reveal the low-field behavior of the linewidth in this temperature region. The connection of these data to a critical line-broadening theory of Huber is discussed. For $T < T_N$ the 5.4-GHz linewidth variation is significantly different from that reported at higher frequencies. Also, for $T < T_N$ the magnetic resonance field at fixed frequency depends on temperature. This effect has not been reported at higher frequencies.

I. INTRODUCTION

This article reports measurements of the paramagnetic and antiferromagnetic resonance linewidth of rubidium manganese flouride ($RbMnF_3$) at *C* band (5.4 GHz). Our results extend earlier measurements by Siebert[1] at 30 GHz and more recently by Gupta and Seehra[2] which were performed at 24.4 and 9.2 GHz. The 5.4-GHz frequency used in the measurements reported here is the lowest frequency (and correspondingly the lowest magnetic field) yet reported for measurements on $RbMnF_3$ near its antiferromagnetic transition temperature. Most of the data fall into one or the other of two categories corresponding, respectively, to $T > T_N$ and $T < T_N$. The $T > T_N$ data provide an additional test of a model by Huber[3] which explains the temperature dependence of the electron paramagnetic resonance (EPR) linewidth of *S*-state ions in insulators in the critical region ($T > T_N$). No theory exists for the second type of data ($T < T_N$). However, these data extend experimental results obtained by Gupta and Seehra[2] and establish that the antiferromagnetic resonance linewidth for temperatures just below T_N has a temperature dependence at 5.4 GHz which is markedly different from that at 9.2 GHz and higher. Finally, we show a limited amount of data for the temperature dependence of the resonance field (H_0) for temperatures just below T_N. This phenomenon appears to exist only at 5.4 GHz and not at the higher frequencies.

In Sec. II the experimental procedures used are described. The data obtained are presented in Sec. III and discussed in Sec. IV.

II. EXPERIMENTAL PROCEDURE

The 5.4-GHz spectrometer used had a simple tee configuration like that described by several authors,[4,5] and had bolometer detection with field modulation at frequencies between 40 and 80 Hz. The $7 \times 4 \times 2$ mm^3 sample weighed 73 mg and was glued to the waveguide wall one-quarter wavelength from the end of the shorted waveguide. To avoid line-broadening effects[6] no microwave cavity was used. The sample used was a portion of the sample Gupta and Seehra used for the earlier 9.2-GHz experiments.[2] The sample was oriented by x-ray Bragg scattering.

The sample temperature was controlled by enclosing the sample and waveguide in an evacuated can and heating with a heater coil which was wrapped around the waveguide. Helium gas was used to thermally link the sample to the surrounding nitrogen bath. The sample temperature was measured with a copper-constantan thermocouple embedded in the wall of the waveguide near the sample. The precision of temperature measurement is estimated to be ± 0.1 K and its accuracy to be ± 0.5 K.

The precision of the linewidth measurement is estimated to be ± 0.3 Oe and can be judged by looking at the scatter in the data points in Figs. 1 and 2. The accuracy of the linewidth measurement is estimated to be ± 1.0 Oe. This potential systematic error is due primarily to inhomogeneities in the magnetic field combined with the fact that the field calibration instrument (an NMR probe using a water sample) was at a slightly different position in the field from the $RbMnF_3$ sample. This procedure provided the advantage of improved precision by permitting the simultaneous measurement of the magnetic field and the microwave absorption due to ESR. In addition to calibrating the magnetic sweep with an NMR probe a computer-assisted linewidth determination procedure was developed which permitted use of the entire line to locate the position of the absorption derivative peaks. Using the entire line for linewidth measurement made the linewidth determination less dependent on small noise bumps and also permitted the Lorentzian character of each line to be verified. The

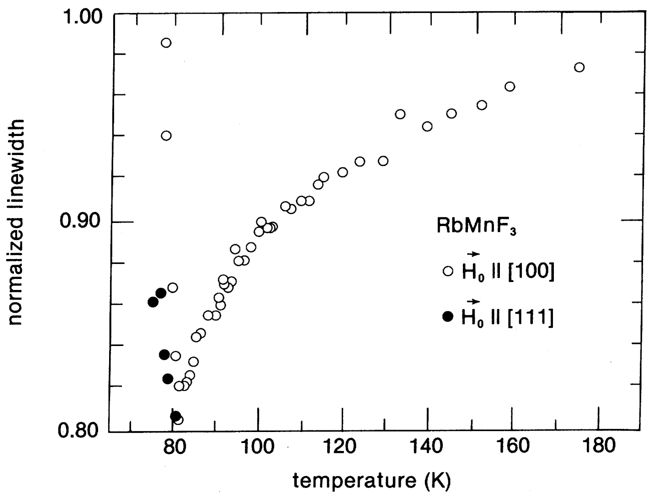

FIG. 1. Temperature dependence of the paramagnetic resonance linewidth at 5.4 GHz. Data for $T > T_N$ and $\vec{H}_0 \parallel [111]$ are not shown since they were judged to be identical within the experimental error to the $T > T_N$, $\vec{H}_0 \parallel [100]$ data shown. The linewidth data are normalized to a room-temperature linewidth of 57.5 Oe.

linewidths reported are the peak-to-peak absorption derivative widths. The spectrometer was tuned using the condition that the two peaks be of equal height. An important experimental detail is that below the Néel temperature (T_N) of about 83 K the resonance field shifts rapidly with temperature (see Fig. 3). A small temperature drift while sweeping through the line can have a large effect on the measured linewidth. This effect produces a line broadening

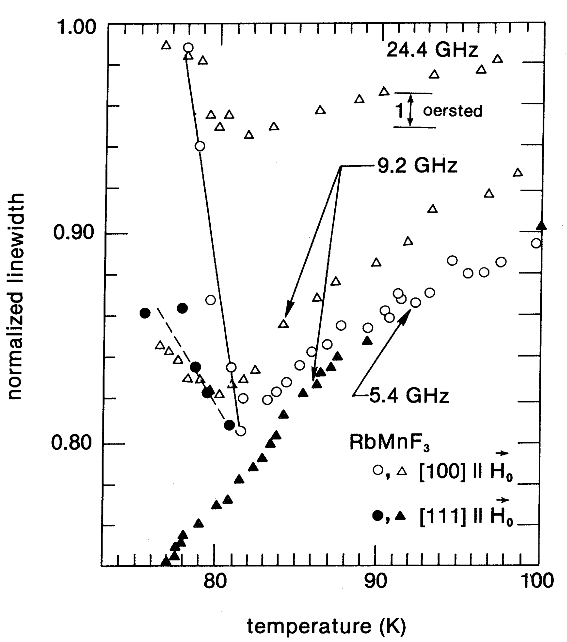

FIG. 2. Comparison of the temperature dependence of the paramagnetic resonance linewidth at 5.4 GHz (this work) with 9.2- and 24.4-GHz data from Ref. 1. The solid line is a fit to the 5.4-GHz, $\vec{H}_0 \parallel [100]$ data for $T < T_N$. The dashed line is a fit to the 5.4-GHz, $\vec{H}_0 \parallel [111]$ data for $T < T_N$.

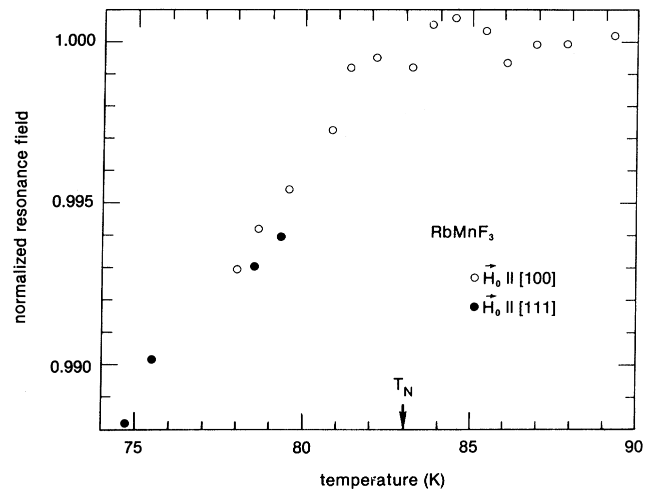

FIG. 3. Temperature dependence of the resonance field at 5.4 GHz. The line center is normalized to its average value for $T > 83$ K.

for one direction of magnetic field sweep and a line narrowing for the other direction assuming a constant direction of temperature drift. Each line was measured twice in succession using each sweep direction in turn, providing a sensitive check on the stability of the temperature.

III. EXPERIMENTAL DATA AND ANALYSIS

The experimental data for the dependence of normalized linewidth on temperature are shown in Fig. 1 for two directions of the magnetic field. Data for $\vec{H}_0 \parallel [100]$ are shown both for temperatures below and above the antiferromagnetic transition temperature, whereas for $\vec{H}_0 \parallel [111]$ only data for temperatures below T_N are shown. The data for $T > T_N$ and $\vec{H}_0 \parallel [111]$ are not shown since it was judged to be identical with the $T > T_N$ data shown within the accuracy of the experiment.

A comparison of this 5.4-GHz data with the 9.2- and 24.4-GHz data from Ref. 2 is shown in Fig. 2. Figure 2 shows a narrower range of temperature than Fig. 1 and emphasizes the temperature region nearer to T_N. In viewing Fig. 2 it should be remembered that at 30 GHz Siebert[2] observed no temperature dependence of the linewidth in the temperature region shown. Two significant features of the data emerge. First, above T_N the 9.2- and 5.4-GHz linewidths have essentially the same temperature dependence. Second, below T_N the 9.2- and 5.4-GHz linewidth graphs behave rather differently. For $\vec{H}_0 \parallel [100]$ the 5.4-GHz line broadens much more rapidly than the 9.2-GHz line, the slopes being -2.9 and -1.0 Oe K^{-1}, respectively. Also, for $\vec{H}_0 \parallel [111]$ the 5.4-GHz line broadens as T is lowered below T_N, whereas the 9.2-GHz line narrows.

The 5.4-GHz data for $T > T_N$ was compared with the theoretical expression

$$\Delta H = A + B(T - T_N)^{\alpha} ,$$

which is derived by Huber.[3] Huber's theory suggests $\alpha = 1.05$. α determined from the 5.4-GHz data is found to have two distinct values, these being $\alpha = 1.6$ for

$T_N < T < 86$ K and $\alpha = 0.8$ for $T > 86$ K.

The temperature dependence of the resonance field (H_0) is shown in Fig. 3. The temperature shift of the normalized line center for $T < T_N$ occurs at the rate 1.5×10^{-3} K^{-1} (for the line center normalized to its room-temperature value). This corresponds to a line displacement rate of 2.9 Oe K^{-1} for a resonance field of 1944 Oe. The data suggest that this rate is the same for the [100] and [111] directions for the temperature range shown.

IV. DISCUSSION

The main conclusion of this paper is that reducing the paramagnetic resonance frequency from 9.2 to 5.4 GHz does not further enhance the line-narrowing effect observed at 9.2 GHz. This conclusion is based on the data for $T > T_N$ shown in Fig. 2. We can say that the "zero-field paramagnetic linewidth" has been observed both in these 5.4-GHz experiments and in the earlier 9.2-GHz experiments of Gupta and Seehra for temperatures above the Néel temperature.

The data for $T > T_N$ bear on a theory developed by Huber[3] to explain the very different temperature dependence of the EPR linewidth (ΔH) in cubic systems ($RbMnF_3$, $KMNF_3$) and uniaxial systems (MnS, MnF_2). Seehra and Huber have reviewed these systems.[7] To summarize their behavior, consider the situation in which the temperature approaches T_N from above. In uniaxial systems such as MnF_2 as $T \rightarrow T_N$ from above, H increases by several orders of magnitude, whereas in cubic systems such as $RbMnF_3$ it is observed to remain constant or even to narrow slightly (see Fig. 1.) The theory of Huber draws upon the combined effects of crystal symmetry and dipolar coupling as the origin of these very different behaviors. The value of α determined by this experiment leaves the question of agreement between experiment and Huber's theory unresolved.

In contrast to the $T > T_N$ case for $T < T_N$ no theory currently exists. Here, the 5.4-GHz data differ significantly from the 9.2-GHz data. The sharpest difference is for the case $\vec{H}_0 \parallel [111]$, where the 5.4-GHz line broadens as T falls below T_N, whereas the 9.2-GHz line continues to narrow. Also, for $\vec{H}_0 \parallel [100]$ the 5.4-GHz line broadens more rapidly than the 9.2-GHz line.

The resonance data for $T < T_N$ correspond to antiferromagnetic resonance (AFMR). The associated domain structure has been studied by Ince and co-workers.[8–10] Studies by Eastman[11] of AFMR in $RbMnF_3$ with uniaxial stress using a 23.2-GHz spectrometer show that the AFMR linewidth is determined by inhomogeneous stress at low temperatures but that an intrinsic relaxation process controls the linewidth at the Néel temperature. No temperature dependence of the linewidth near T_N was mentioned.

Previous work bearing directly on the problem of magnetic-field-dependent relaxation just below T_N in antiferromagnets is not extensive. Seehra and Huber[7] review the relaxation process near critical points in magnetic insulators, but their discussion applies to the paramagnetic regime. Rezende and White[12] have done detailed theoretical investigations of multimagnon relaxation in $RbMnF_3$ as the temperature increases toward T_N, but emphasized a lower temperature region and zero magnetic field.

Turning to a discussion of the temperature dependence of the resonance field for $RbMnF_3$ for fixed frequency, no previous investigators[1,2,10,11] report any temperature dependence of H_0, and one[1] implies that H_0 was temperature independent down to 80.4 K within the accuracy of his experiment (about 1%). Our data indicate significant shifts in H_0 exceeding 1% of the room-temperature value below 76 K and appearing to be linear with temperature. In conclusion, in $RbMnF_3$ the temperature dependence of the linewidth and of the resonance field at fixed frequency just below T_N changes significantly when the resonance frequency is decreased from 9.2 to 5.4 GHz, but above T_N the behavior is the same at these two frequencies.

ACKNOWLEDGMENTS

We wish to thank Mr. Ronald Huinker for his contribution to the construction and testing of the spectrometer used in this experiment, and to Professor Richard Barnes of Iowa State University for the loan of C-band microwave components. We especially thank Dr. Mohindar Seehra for useful conversations, and also for providing the sample used in these experiments. This research was supported in part by a grant from the Research Corporation.

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*Permanent address: Department of Physics, University of Chicago, Chicago, IL 60637.

[1]J. F. Siebert, Ph.D. thesis, University of California at Berkeley, 1968.

[2]R. P. Gupta and M. S. Seehra, Phys. Lett. 33A, 347 (1970).

[3]D. L. Huber, Phys. Lett. 37A, 283 (1971).

[4]M. S. Seehra, Ph.D. thesis, The University of Rochester, 1969 (unpublished).

[5]R. C. Nicklin, Ph.D. thesis, Iowa State University, 1967 (unpublished).

[6]M. S. Seehra, Rev. Sci. Instrum. 39, 1044 (1968).

[7]M. S. Seehra and D. L. Huber, in *Magnetism and Magnetic Materials—1974*, edited by C. D. Graham, Jr., G. H. Lander, and J. J. Rhyne, AIP Conf. Proc. No. 24 (AIP, New York, 1975), pp. 261–267.

[8]W. J. Ince, J. Appl. Phys. 37, 1132 (1966).

[9]W. J. Ince and A. Platzker, Phys. Rev. 175, 650 (1968).

[10]W. J. Ince, Phys. Rev. 184, 574 (1969).

[11]D. E. Eastman, Phys. Rev. 156, 645 (1967).

[12]S. M. Rezende and R. M. White, Phys. Rev. B 18, 2346 (1978).